Characterization and source apportionment of atmospheric organic and elemental carbon during fall and winter of 2003 in Xi'an, China

J. J. Cao, J. C. Chow, S. C. Lee, Y. Li, S. W. Chen, Z. S. An, K. Fung, J. G. Watson, C. S. Zhu, S. X. Liu

Atmos. Chem. Phys. Discuss., 5, 3561–3593, 2005
www.atmos-chem-phys.org/acpd/5/3561/
SRef-ID: 1680-7375/acpd/2005-5-3561
European Geosciences Union

Atmospheric Chemistry and Physics Discussions

Characterization and source apportionment of atmospheric organic and elemental carbon during fall and winter of 2003 in Xi'an, China

J. J. Cao[1], J. C. Chow[2], S. C. Lee[3], Y. Li[1], S. W. Chen[4], Z. S. An[1], K. Fung[5], J. G. Watson[2], C. S. Zhu[1], and S. X. Liu[1]

[1]State Key Laboratory of Loess & Quaternary Geology, Institute of Earth Environment, Chinese Academy of Sciences, Xi'an 710075, China
[2]Desert Research Institute, Reno, USA
[3]The Hong Kong Polytechnic University, Hong Kong, China
[4]Tongji University, Shanghai 200 092, China
[5]AtmAA Inc., Calabasas, CA, USA

Received: 14 March 2005 – Accepted: 7 April 2005 – Published: 1 June 2005

Correspondence to: J. J. Cao (cao@loess.llqg.ac.cn)

Abstract

Continuous observation of atmospheric organic and elemental carbon (OC, EC) were conducted at Xi'an during high pollution seasons from September 2003 to February 2004. $PM_{2.5}$ samples were collected on pre-fired quartz-fiber filters with battery-powered mini-volume samplers every day and PM_{10} samples were collected every third days. Three types of source samples (i.e., coal-combustion, motor vehicle exhaust, and biomass burning) were also collected during ambient sampling period. Ambient and source samples were analyzed for OC and EC by thermal/optical reflectance (TOR) following the Interagency Monitoring of Protected Visual Environments (IMPROVE) protocol. The average $PM_{2.5}$ OC concentrations in fall and winter were 34.1±18.0 μg m^{-3} and 61.9±33.2 μg m^{-3}, respectively, while EC were 11.3±6.9 μg m^{-3} and 12.3±5.3 μg m^{-3}, respectively. Most of OC and EC were associated with fine particle ($PM_{2.5}$) mode. The OC and EC levels at Xi'an are higher than most urban cities in Asia. The OC and EC in fall were found to be strongly correlated (R^2>0.9), with moderate correlation in winter (R^2=0.66). The carbonaceous aerosol accounted for 48.8±10.1% of the $PM_{2.5}$ during fall and 45.9±7.5% during winter. Average OC/EC ratio was 3.3 in fall and 5.1 in winter with individual OC/EC ratios constantly exceeding 2.0. Elevated OC/EC ratios were found during heating seasons with increased coal combustion. The contribution of secondary organic carbon was not significant during winter. The time series of OC and EC showed periodic variability. Traffic contributes 5 and 7 day peaks in the spectrum, precipitation appears as a 10 day periodicity and biomass burning can be identified as a 24 day periodicity. Total carbon (TC) was apportioned by absolute principal component analysis (APCA) using the 8 carbon fraction data (OC1, OC2, OC3, OC4, EC1, EC2, EC3, and OP [a pyrolyzed carbon fraction]). TC attributes 73% to gasoline exhaust, 23% to diesel exhaust, and 4% to biomass burning during fall. However, TC attributes 44% each to gasoline exhaust and coal burning, 9% to biomass burning, and 3% to diesel exhaust during winter.

ACPD

5, 3561–3593, 2005

1. Introduction

Study of atmospheric organic carbon (OC) and elemental carbon (EC, also named black carbon, BC) in China has received intensive attentions in recent years due to the high emission of OC and EC and their impacts on global/regional climatic and environmental changes (IPCC, 2001; ACE-Asia, 1999, http://saga.pmel.noaa.gov/aceasia/; Project Atmospheric Brown Clouds (ABC), 2003, http://www-abc-asia.ucsd.edu/). Roughly one-fourth of global BC emissions are estimated from China (Cooke et al., 1999). Control of emission of BC from China was considered as a most effective way for slowing global warming (Jacobson et al., 2002). Elevated OC and EC contributed to high particulate matter (PM) pollution in urban area (He et al., 2001; Ye et al., 2003; Cao et al., 2003, 2004). In a regional scale, EC heats the air, alter atmospheric stability and vertical motions, and affect the large-scale circulation and hydrologic cycle with significant regional climate effects in China (Menon et al., 2002).

Several studies have been conducted on developed and coastal cities like Beijing, Shanghai, Guangzhou, and Hong Kong (He et al., 2001, Ye et al., 2003; Cao et al., 2003, 2004), but limited studies were available for inland cities in China. Xi'an situated in northwest China and it was selected as the city with high PM pollution in the United Nation Development Program (UNDP) pilot study during 1997 and 2001 (Zhang et al., 2001, 2002). Xi'an has been the capital city of 13 Chinese dynasties for more than a millennium. Since the discovery of terra-cotta warriors and horse, Xi'an became one of the most popular tourist attractions in China. In addition to coal-combustion and vehicle exhaust, Xi'an is impacted by mineral dust and dust storm (Zhuang et al., 1992, 1993; Gao et al., 1997; Cao et al., 2005). Therefore, the primary objectives of the paper are to: 1) examine the temporal variations of $PM_{2.5}$ (particle diameter smaller than 2.5 μm) and PM_{10} (particle diameter smaller than 10 μm) OC and EC concentrations, and 2) quantify the contributions of major sources to OC and EC.

ACPD
5, 3561–3593, 2005

2. Sampling and analysis

2.1. Sampling sites and descriptions

Xi'an (33°29′–34°44′ N, 107°40′–109°49′ E, population 5 million), the largest city in northwestern China, lies in the south margin of the Loess Plateau, China. The Xi'an monitoring site (400 m above the sea level) (Fig. 1) is situated ~15 km south of downtown Xi'an. The site has an urban-scale zone of representation (Chow et al., 2002) with no major industrial activities or local fugitive dust sources in the surrounding area, which is mostly residential. $PM_{2.5}$ and PM_{10} samples were obtained from the rooftop of the Institute of Earth Environment, Chinese Academy of Sciences, at 10 m above ground level. PM samples were collected during fall (13 September 2003 to 31 October 2003) and winter (1 November 2003 to 29 February 2004).

2.2. Sample collection

Daily $PM_{2.5}$ and every third days PM_{10} samples were collected using two battery-powered mini volume samplers (Airmetrics, Oregon, USA) operating at flow rates of 5 / min^{-1} (Cao et al., 2003). Prior to field operations, calibrated mini-vol samplers were collocated with low volume $PM_{2.5}$ and PM_{10} Partisol samplers (model 2000, Rupprecht & Patashnick, Albany, New York, USA) at The Hong Kong Polytechnic University for data comparison. The difference between the two types of samplers was less than 5% for the $PM_{2.5}$ and PM_{10} mass.

PM samples were collected on 47 mm Whatman quartz microfiber filters (QM/A); the filters were pre-heated at 900°C for 3 h before sampling. The exposed filters were stored in a refrigerator at about 4°C before chemical analysis to prevent the evaporation of volatile components. Quartz-fiber filters were analyzed gravimetrically for mass concentrations using a Sartorius MC5 electronic microbalance with a ±1 μg sensitivity (Sartorius, Göttingen, Germany). These filters were weighed after 24-h equilibration at temperature between 20°C and 23°C and relative humidity (RH) between 35% and

ACPD
5, 3561–3593, 2005

45%. Each filter was weighed at least three times before and after sampling, and the net mass was obtained by subtracting the difference between the averaged pre- and post-sampling weights. Precisions of the weighings were <10 μg for blank filters and <20 μg for filter samples. A total of 16 field blanks were collected to subtract the positive artifacts due to adsorption of gas-phase organic components onto the filter during and/or after sampling. However, negative artifacts due to volatilization of particle-phase organics from particle sample were not quantified. A total of 165 $PM_{2.5}$ and 53 PM_{10} samples were collected, respectively. Five $PM_{2.5}$ source samples were collected from the coal-combustion of residential stoves, six samples collected from a major highway with heavy traffic, and five samples collected from a farmland when the maize residue combusted after harvest during ambient sampling period.

Continuous meteorological data were monitored by HFY-IA Wind Speed/Wind Direction Instrument (Changchun Institute of Metrological Instruments, Changchun, Jilin Province, China).

2.3. Thermal/optical carbon analysis

The samples were analyzed for OC and EC using a Desert Research Institute (DRI) Model 2001 Thermal/Optical Carbon Analyzer (Atmoslytic Inc., Calabasas, CA, USA). A 0.5 cm^2 punch from the filter was analyzed for eight carbon fractions following the IMPROVE (Interagency Monitoring of Protected Visual Environments) thermal/optical reflectance (TOR) protocol (Chow et al., 1993, 2001, 2004; Fung et al., 2002). This produced four OC fractions (OC1, OC2, OC3, and OC4 at 120°C, 250°C, 450°C, and 550°C, respectively, in a He atmosphere); a pyrolyzed carbon fraction (OP, determined when reflected a transmittance laser light attained its original intensity after O_2 was added to the analysis atmosphere); and three EC fractions (EC1, EC2, and EC3 at 550°C, 700°C, and 800°C, respectively, in a 2% O_2/98% He atmosphere). IMPROVE OC is operationally defined as OC1+OC2+OC3+OC4+OP and EC is defined as EC1+EC2+EC3-OP. Inter-laboratory comparisons of samples between IMPROVE protocol with the DRI Model 2001 instrument and the TMO (thermal manganese diox-

ACPD
5, 3561–3593, 2005

ide oxidation) method (done by AtmAA, Inc., Calabasas, CA) has shown difference <5% for total carbon (TC) and 10% for OC and EC (Fung et al., 2002). Average field blanks were 1.56 and 0.42 μg m^{-3} for OC and EC, respectively. Quality Assurance/Quality Control (QA/QC) procedures were described in Cao et al. (2003).

3. Results and discussion

3.1. Temporal variations of OC and EC

Monthly and seasonally averaged OC and EC concentrations are summarized in Table 1. $PM_{2.5}$ OC and EC during winter are 1.8 and 1.1 times, respectively, of those during fall. PM_{10} OC and EC during winter are 2.2 times and 1.5 times, respectively, of those during fall. Monthly average OC and EC were highest during December and lowest during September. In December OC in $PM_{2.5}$ and PM_{10} were 81.7±36.2 μg m^{-3} and 124.8±54.8 μg m^{-3}, respectively, and EC in $PM_{2.5}$ and PM_{10} were 15.2±4.6 μg m^{-3} and 28.9±8.9 μg m^{-3}, respectively. For $PM_{2.5}$ and PM_{10}, highest to lowest values of OC were 3.3 and 4.2 and EC were 1.8 and 2.6, respectively. High variability of OC concentrations may be due to the contributions of different emission sources.

Figure 2 shows that temporal variations of $PM_{2.5}$ OC coincide with mass, and to a lesser extend, with EC. The Pearson correlation coefficients of these two series was as high as 0.96 (significant level 99%), pointing to OC is major contributor to $PM_{2.5}$ mass. The Person correlation coefficients of $PM_{2.5}$ mass and EC also reached to 0.72 (significant level 99%), implying EC is a significant contributor to $PM_{2.5}$ mass. $PM_{2.5}$ OC increased gradually from September to November, and reached a maximum on 14 December 2003 (189.6 μg m^{-3}). The PM variations at Xi'an have a winter maximum and summer minimum (Zhang et al., 2001), so this OC value is expected to be the highest value in the year. $PM_{2.5}$ OC concentrations fluctuated from mid-December to earlier January. After the Chinese Spring Festival (22 January 2004 to 29 January 2004), OC dropped to a low level and began to decrease during February. Similar trend

ACPD
5, 3561–3593, 2005

of EC variation was found, but the EC concentration was lowest during the Chinese Spring Festival and fluctuated in low values from 22 January 2004 to 5 February 2004.

The major emission sources of OC and EC in China includes coal-combustion, motor vehicle exhaust, and biomass burning (includes biofuels) (Streets et al., 2001). Previous study showed that coal-combustion and motor vehicle exhaust were the two major emission sources of TSP (Total Suspended Particle) (Zhang et al., 2001). During the study periods, the impact of biomass burning seems evident because Xi'an is located in the Guanzhong Plain, a major base of national rice/corn production. During the fall harvest season in mid-October, the residue of diverse crops like corn were mainly cleaned by field combustion. Residential combustion of biofuels is also a significant contributor during wither because part of farmer around Xi'an use it as heating source. Therefore, coal-combustion, motor vehicle exhaust, and biomass burning constitute the major emission sources of carbonaceous pollution during fall and winter in Xi'an.

Wet deposition is the dominant mechanism to remove OC and EC. As shown in Table 2, the precipitation days in fall were 13 days, account for 27% of observational days, however, compared to 6 days or 5% of total observational days in winter. During precipitation, average OC and EC decreased to 16.2 μg m^{-3} and 4.3 μg m^{-3}, respectively, which may mirror the urban background values of OC and EC in high pollution seasons at Xi'an. Average OC and EC levels in winter precipitation days were two times of those in fall, i.e., with 34.7 μg m^{-3} for OC and 8.6 μg m^{-3} for EC, reflecting the low quantity and shorter duration of precipitation in winter. $PM_{2.5}$ mass and carbon concentrations increased by two to three folds during normal day period for both seasons.

3.2. Relationship between OC and EC

The origin of carbonaceous particles can be estimated on the basis of the relationship between OC and EC (Turpin and Huntzicker, 1991; Chow et al., 1996). As shown in Fig. 3, strong OC-EC correlations (0.90–0.95) were found in fall, suggesting impacts from similar sources (e.g., motor vehicle exhaust and biomass burning). In contrast, the correlations (0.66) were low in winter, pointing to the complex of emission sources.

Coal-combustion from residential heating in winter is the major emission sources of OC and EC. It contributes to more than 50% to TSP (1997 data, Zhang et al., 2001). Even though many residents in Xi'an replaced coal by natural gas, a large body of low-income families still use coal as major source of cooking and heating in winter. Coal-combustion boilers has been banned within the second beltway in the downtown since 1998, but many middle- and small- scales coal-combustion boilers inside and outside Xi'an downtown are still in use due to its low cost.

The slopes of OC versus EC in winter were high, 5.12 for $PM_{2.5}$ and 3.83 for PM_{10}, as compared to those in fall (2.46) (Fig. 3), implying the emission of OC increased largely relative to EC in winter. The difference may be ascribed to the change of emission sources in two seasons.

3.3. Variability of OC/EC ratios

Atmospheric EC originates from primary anthropogenic sources and is not formed by reactions involving gaseous hydrocarbon precursors in the atmosphere. OC may be emitted directly from sources as primary particles, but secondary organic aerosols (SOA) can also be formed in the atmosphere from the low vapor pressure products by atmospheric chemical reactions. The ratio of OC to EC concentrations has been used to study emission and transformation characteristics of carbonaceous aerosol.

As shown in Table 1, average OC/EC ratios in $PM_{2.5}$ and PM_{10} ranged from 3.0 to 3.4 in fall, and increased to 3.6–6.4 in winter. Monthly averages of OC/EC ratios ranged 3.3–6.4 in $PM_{2.5}$ and 3.0–5.1 in PM_{10} with the highest ratios found in January.

Daily variations of $PM_{2.5}$ OC/EC ratios in Fig. 2 showed lower ratios and less variations in fall and higher ratios and more variations in winter. The ratios were around 8.0 during 13 December 2003 to 15 December 2003, and reached a peak value of 9.0 during 22 January 2004 to 29 January 2004. OC/EC ratios were normally affected by three factors: 1) emission sources; 2) the formation of SOA in the atmosphere; 3) the removal of OC and EC. Since EC has higher washout efficiency than OC (Cachier et al., 1996), OC/EC ratio can be increased accordingly. From Table 2, average OC/EC

ACPD
5, 3561–3593, 2005

ratios were 4.0 under precipitation days. Since the precipitation days were small (only 5%) in winter (116 days), the effect of OC/EC ratios during and 1–2 days after precipitation are expected to be low. A long precipitation period during Chinese National Holiday (from 28 September 2003 to 4 October 2003) led to a peak OC/EC ratio of 6.0 in fall.

The OC/EC ratios exceeding 2.0 have been used to indicate the presence of SOA (Gray et al., 1986; Chow et al., 1996). Pandis' study (Pandis, 2002) indicated that the formation of SOA in the ambient air was mainly controlled by temperature. The temperature at Xi'an ranged from −5°C to 5°C with dense haze and short duration of sunshine in winter. So the formation of SOA in Xi'an may be low. Cabada's study (Cabada, 2002) in western Pennsylvania also confirmed that the contribution of SOA in winter decreased to zero.

Average OC/EC ratio were 12.0 in coal-combustion samples, 4.1 in motor vehicle (from gasoline vehicles) exhaust samples, and 60.3 in biomass burning samples. For comparison, the OC/EC ratio was 2.7 for coal-combustion sample and 1.1 for motor vehicle sample (Watson et al., 2001), and 9.0 for biomass burning (Cachier et al., 1989). The individual OC/EC ratios for this study exceeded 2.0 for both $PM_{2.5}$ and PM_{10} fractions (Fig. 2), which may mirror the joint contributions from coal-combustion, motor vehicle exhaust, and biomass burning sources. Elevated OC/EC ratios (8.0) during 12 December 2003 to 14 December 2003 can be attributed to biomass burning. High OC/EC ratios (6.0–9.0) during the Chinese Spring Festival (22 January 2004 to 5 February 2004) was owing to less contribution from motor vehicle during holiday and more contribution from residential coal-combustion.

3.4. Contributions to $PM_{2.5}$ and PM_{10} mass

The box plot in Fig. 4 showed that PM_{10} was more scatter than $PM_{2.5}$ in both seasons. Daily PM_{10} in winter varied by a factor of 5.7, ranging from 155 μg m^{-3} (6 November 2003) to 885 μg m^{-3}(14 December 2003). The average $PM_{2.5}$ was 140.1 μg m^{-3} in fall

ACPD
5, 3561–3593, 2005

and 258.7 μg m^{-3} in winter. The average PM_{10} was 261.9 μg m^{-3} in fall and 450.6 μg m^{-3} in winter. $PM_{2.5}$ accounted for 55.6% of the PM_{10} in fall, ranging between 44.3% and 77.4%. In contrast, $PM_{2.5}$ accounted for 60.4% of the PM_{10} in winter with a large range from 33.0% and 97.6%. For comparison, the order of the percentage of $PM_{2.5}$ in the PM_{10} in Chinese urban cities was Shenzhen (73.3%, 2001) (Cao et al., 2003) > Zhuhai (70.8%, 2001) (Cao et al., 2003) > Chongqing (65.1%, 1997) (Wei et al., 1999) > Wuhan (60.5%, 1997) (Wei et al., 1999) > Xi'an (60.4%, 2003) > Lanzhou (51.9%, 1997) (Wei et al., 1999). In reference to Class 2 of the Chinese PM_{10} standard (150 μg m^{-3}) (GB 3905–1996), only 5 out of the PM_{10} sampling days in fall in compliance with the legislation and none of the days in winter. This implies a serious PM pollution in Xi'an even with substantial efforts on pollution to control by local government.

As shown in Table 3, total carbonaceous aerosol (TCA), the sum of organic matter (OM=1.6×OC) and EC, contributed 48.8% of $PM_{2.5}$ in fall and 45.9% in winter. The percentage of TCA in PM_{10} was lower than $PM_{2.5}$, with an average of 34.5% in fall and 37.0% in winter, may due to an increased contributions of geological matter in coarse particles. Taking into account the contributions of other sources such as geological materials and secondary aerosol (sulfate, nitrate, and ammonium) to $PM_{2.5}$, it is reasonable to speculate that TCA is the most important contributor to fine particles. The material balance showed that TCA is the dominant components of $PM_{2.5}$ (Li Y., 2004). It is thus that control measures should be aimed to the anthropogenic combustion sources rather than fugitive dust.

From time series in Fig. 2, TCA% varied around the 45% level during the six months and it didn't correlate with the changes of $PM_{2.5}$ mass or the OC and EC concentrations. Level of TCA% is co-varied with the precipitant events and low PM periods. During precipitation, OC and EC usually resided in sub-micron size ranges (0.01–1 μm), leading to an increase in the percentage of OC and EC and resulted in elevated TCA% levels.

$PM_{2.5}$ OC accounted for 81.8% and 72.8%, respectively, of PM_{10} OC, whereas $PM_{2.5}$ EC accounted for 75.0% and 59.6% of PM_{10} EC in fall and winter, respectively (Ta-

ACPD
5, 3561–3593, 2005

ble 1). Less than 60% of PM_{10} EC resided in $PM_{2.5}$ in winter, reflecting the presence of coarse soot particles in the emission of incomplete coal-combustion.

3.5. The characterization of eight carbon fractions

The IMPROVE TOR protocol does not advance from one temperature to the next until a well-defined carbon peak has evolved (Chow et al., 1993, 2004). Carbon abundances in each of these fractions differ by carbon source (Chow et al., 2003). 8 carbon fractions have been utilized for the source apportionment of carbonaceous aerosol (Kim et al., 2003a, 2004).

The average percentages of 8 carbon fractions in ambient and source samples are shown in Fig. 5. There are distinct differences among three sources samples for 8 carbon fractions. OC2 accounted for 46.9% of TC in coal-combustion samples, higher than 29.2% in biomass burning samples and 30.5% in motor vehicle samples. OC1 contributed 36.8% to TC in biomass burning samples, higher than 2.0% in coal-combustion samples and 2.8% in motor vehicle exhaust samples. EC1 constituted 15.4% to TC, higher than 5.6% in coal-combustion samples and 0.4% in biomass burning samples.

Monthly variations of the 8 carbon fractions were related to the contributions of different emission sources. November experienced the highest contribution from biomass burning sources, with OC1 attaining 8.7%, which was the highest value in six months. It decreased to 1.7% in February. The variations of OC1 may point to the contributions of biomass burning in different months. OC2 increased in six months (except November), possible reflecting the increased contributions of coal-combustion from fall to winter. EC1 reached its lowest values in January, implying decreased contributions of motor vehicle exhaust. This may be due to the less activity of motor vehicles during the Chinese Spring Festival, consistent with the analysis of OC/EC ratios (Sect. 3.3). OP was 22.1%, 20.8%, 25.3%, 21.4%, 20.5%, and 16.0% in six months, with an average of 21.0%. In contrast, the percentage of OP in TC varied between 8.0% and 17.8% during summer for Pearl River Delta Region in China (Cao et al., 2004).

ACPD
5, 3561–3593, 2005

3.6. Periodic characteristics of OC and EC

Concentrations of OC and EC were influenced by emissions and dilution processes. The periodic features of emission sources and metrological conditions can be identified from the time series of OC and EC. Hies (2000) showed that domestic heating by coal combustion appears with a 365 day periodicity, traffic contributes 3.5, 4.6 and 7 day peaks in the spectrum and elevated long range EC can be identified as characteristic peaks with periodicities in the range from 13 to 42 days in Berlin, Germany.

The comparison of periodicities of OC, EC, $PM_{2.5}$ mass and daily average wind speed are illustrated in Fig. 6. These curves were obtained by AutoSignal 1.0 software (SPSS, USA). The common periodicities of OC, EC and $PM_{2.5}$ were 24 days, 10 days, 7 days and 5 days. The periodicities of OC were same as $PM_{2.5}$, implying they were controlled by similar process, consistent with correlation of OC with $PM_{2.5}$. 7 days and 5 days were the periodicities of motor vehicle variations, in agreement with Hies' study (Hies, 2000). Precipitation events had 10 days periodicity from September to November. This periodicity should reflect the impact of precipitation on OC and EC concentrations. 24 days periodicity may point to the biomass burning events since the biomass burning events occurred about every 24 days. In addition, EC also had components of 60 days and 13 days peaks in the spectrum. 13 days periodicity was a major component in the spectrum of wind speed, which has been identified by Hies (2000). Wind speed also influences EC concentrations. 60 day peaks may be related to the change of primary emission sources. From Fig. 2, September and October were the low period of EC, November and December were the high period of EC, January and February were also the low period of EC. In conclusion, 7 days and 5 days periodicities of OC and EC were controlled by motor vehicle exhaust, 10 days periodicities were related to precipitation, and 24 days periodicities were associated with the biomass burning around Xi'an.

ACPD
5, 3561–3593, 2005

3.7. Comparison of OC and EC with other Asian cities

At least 30 analytical methods existed for OC and EC in different labs, TC measures agree (Schmid et al., 2001) but differences can be found for OC and EC.

Table 4 lists the TC, OC and EC concentrations from Xi'an and other Asian cities. TC in fall and winter at Xi'an ranked the highest, reflecting serious carbonaceous pollution in Xi'an. Comparing with the data from Beijing, Xi'an OC and EC were same as Beijing's in fall. Xi'an EC was same as Beijing EC in winter, but Xi'an OC was two times of Beijing OC. From the analysis of OC and EC distributions in source samples, high OC contents in Xi'an could be ascribed to contributions from coal-combustion. More motor vehicles and less coal are used in Beijing (Yang et al., 2005). Winter OC levels in Xi'an were 3.6, 6.4, 2.7, 4.7, and 5.1 times than those in Shanghai, Hong Kong, Guangzhou, Shenzhen and Zhuhai. Winter EC levels in Xi'an were 1.5, 2.6, 1.5, 2.0, and 2.5 times those of the coastal cities. The lower difference for EC may be attributed to the high emissions of motor vehicle exhaust in these coast cities and the larger difference of OC may be ascribed to the lower use of coal for residential heating. Winter OC and EC levels in Xi'an were 12.4 and 2.9 times those in Chongju, South Korea (Lee et al., 2001).

3.8. Source apportionment of carbonaceous aerosol

8 carbon fractions of TC have been utilized to estimate sources attributions (Kim et al., 2003a, b, 2004). Absolute principal component analysis (APCA) (Thurston & Spengler, 1985) was used in this study.

The first step in APCA is the normalization of all carbon concentrations as Z_{ik}; this is done through the addition of a zero concentration sample as case 0.

$$Z_{ik} = (C_{ik} - C_i)/S_i \quad (1)$$

where C_{ik} is the concentration of carbon fraction i in sample k, C_i is the arithmetic mean concentration of carbon fraction i, and s_i is the standard deviation of carbon

fraction i for all samples included in the analysis. The normalization process allows any continuous variables, such as wind speed to be included in future analyses along with the carbon data.

Regressing the TC data on these absolute principal component scores (APCS) gives estimates of the coefficients which convert the APCS into TC-contributions from each source for each sample. For each source identified by the APCA, the weighted regression of each carbon's concentration on the predicted TC contributions yields estimates of the content of that carbon in each source, as follows:

$$C_{ik} = b + \sum_{j=1}^{n} a_{ij} M_{jk} \quad (2)$$

where C_{ik} is the concentration of carbon fraction i in sample k; b is a constant; a_{ij} is the mean TC fraction of source j's particles represented by carbon fraction i, and M_{jk} is the TC concentration of source j for observation k. By repeating this weighted least square regression for each of the i=1, 2,...n carbon fractions considered in this analysis, one can estimate the mean concentration of the carbon fractions in each factor.

The statistical results for fall and winter were summarized in Tables 5 and 6.

Factor 1 (F1) in fall was highly loaded with the following carbon fractions: OC2, OC3, OC4, OP, and EC1. This factor appears to represent gasoline motor vehicle exhaust. The second component (F2) was highly loaded with EC2 and EC3 and appears to represent diesel vehicle exhaust because it contains lots of high temperature component of EC particles (Watson et al., 1994). The high loading of OC1 in the factor 3 (F3) reflects the contribution of biomass burning. The interpretation of the first factor (F1) in winter is complicated because it is highly loaded with OC2, OC3, OC4, and EC1. This factor may represent the mixture of coal-combustion and motor vehicle exhaust because these two sources are correlated in winter by similar dispersion conditions for surface-based emissions. Similar to the fall results, F2 and F3 in winter represents biomass burning and diesel vehicle exhaust, respectively.

ACPD
5, 3561–3593, 2005

To simplify the estimation, we assumed there is no contribution of coal-combustion in fall and there are contributions from gasoline motor vehicle in fall and winter. The contribution of winter F1 to TC subtracted from the contribution of fall F1 was considered as the contribution of coal-combustion to TC. Thus the source attributions can be resolved for the two seasons (Fig. 7). TC is composed of 73% from gasoline exhaust, 23% from diesel exhaust, and 4% from biomass burning during fall. TC during winter receives 44% from gasoline exhaust, 44% from coal burning, 9% from biomass burning, and 3% from diesel exhaust. The TC source apportionments during fall and winter were consistent with the qualitative analysis of different sources in aforementioned sections.

4. Conclusions

Six-month continuous observations of OC and EC were conducted at Xi'an to gain insight into the characterization and source apportionment of OC and EC. Major findings are as follows.

1) Average $PM_{2.5}$ OC concentrations during fall and winter were 34.1±18.0 μg m^{-3} and 61.9±33.2 μg m^{-3}; and EC concentrations were 11.3±6.9 μg m^{-3} and 12.3±5.3 μg m^{-3}, respectively. Carbonaceous aerosol accounted for 48.8±10.1% and 45.9±7.5% of $PM_{2.5}$ and 34.5±9.3% and 37±8.9% of PM_{10} during fall and winter, respectively. This indicates that carbonaceous aerosol is the dominant component of fine particles in a typical city with high mineral dust pollution in north China like Xi'an.

2) All the OC/EC ratios exceeded 2.0 and average OC/EC ratio were 3.3 in fall and 5.1 in winter. Elevated OC/EC ratios were found during heating seasons with increased primary emission sources like coal combustion. $PM_{2.5}$ OC and PM_{10} OC were highly correlated (R^2=0.90–0.95) during fall, and moderately correlated (R^2=0.66) during winter.

3) $PM_{2.5}$ TC source apportionment by APCA attributes 73% to gasoline exhaust, 23% to diesel exhaust, and 4% to biomass burning during fall. $PM_{2.5}$ TC source apportionment attributes 44% each to gasoline exhaust and coal burning, 9% to biomass

ACPD
5, 3561–3593, 2005

burning, and 3% to diesel exhaust during winter. Therefore, motor vehicle exhaust and coal-combustion were the dominant sources for carbonaceous aerosol in Xi'an, which should be paid more attentions to control them.

Acknowledgements. This project is supported by the National Natural Science Foundation of China (40205018, 40121303), National Basic Research Program of China (2004CB720203), and Research Grants Council of Hong Kong (PolyU5038/01E, PolyU5145/03E), Area of Strategic Development on Atmospheric and Urban Air Pollution (A516) funded by The Hong Kong Polytechnic University.

References

Cabada, J. C., Pandis, S. N., and Robinson, A. L.: Sources of Atmospheric Carbonaceous Particulate Matter in Pittsburgh, Pennsylvania, J. Air & Waste Manage. Assoc., 52, 732–741, 2002.

Cachier, H., Liousse, C., Pertuisot, M. H., et al.: African fire particulate emissions and atmospheric influence, in: Biomass Burning and Global Change, edited by: Levine, J. S., 428–440, MIT Press, Cambridge, Mass., 1996.

Cachier, H., Bremond, M. P., and Buat-Menard, P.: Carbonaceous aerosols from different tropical biomass burning sources, Nature, 340, 371–373, 1989.

Cao, J. J., Lee, S. C., Ho, K. F., et al.: Characteristics of Carbonaceous Aerosol in Pearl River Delta Region, China during 2001 Winter Period, Atmos. Environ., 37, 1451–1460, 2003.

Cao, J. J., Lee, S. C., Ho, K. F., et al.: Spatial and seasonal variations of atmospheric organic carbon and elemental carbon in Pearl River Delta Region, China, Atmos. Environ., 38, 4447–4456, 2004.

Cao, J. J., Lee, S. C., Zhang, X. Y., et al.: Characterization of airborne carbonate over a site on Asian Dust source regions during 2002 spring and its climatic and environmental significance, J. Geophys. Res., 110, D03203, doi:10.1029/2004JD005244, 2005.

Chow, J. C., Watson, J. G., Pritchett, L. C., et al.: The DRI thermal/optical reflectance carbon analysis system: description, evaluation and applications in US air quality studies, Atmos. Environ., 27A (8), 1185–1201, 1993.

Chow, J. C., Watson, J. G., Lu, Z., et al.: Descriptive analysis of $PM_{2.5}$ and PM_{10} at regionally representative locations during SJVAQS/AUSPEX, Atmos. Environ., 30(12), 2079–2112, 1996.
Chow, J. C., Watson, J. G., Crow, D., et al.: Comparison of IMPROVE and NIOSH Carbon Measurements, Aerosol Sci. Technol., 34, 23–34, 2001.
Chow, J. C., Engelbrecht, J. P., Watson, J. G., et al.: Designing monitoring networks to represent outdoor human exposure, Chemosphere, 49(9), 961–978, 2002.
Chow, J. C., Watson, J. G., Kuhns, H. D., et al.: Source profiles for industrial, mobile, and area sources in the Big Bend Regional Aerosol Visibility and Observational (BRAVO) Study, Chemosphere, 54(2), 185–208, 2003.
Chow, J. C., Watson, J. G., Chen, L. W., et al.: Equivalence of elemental carbon by thermal/optical reflectance and transmittance with different temperature protocols, Environ. Sci. Technol., 38(16), 4414–4422, 2004.
Cooke, W. F., Liousse, C., Cachier, H., et al.: Construction of a $1^\circ \times 1^\circ$ fossil fuel data set for carbonaceous aerosol and implementation and radiative impact in the ECHAM4 model, J. Geophys. Res., 104, 22 137–22 162, 1999.
Dan, M., Zhuang, G. S., Lia, X. X., et al.: The characteristics of carbonaceous species and their sources in $PM_{2.5}$ in Beijing, Atmos. Environ., 38, 3443–3452, 2004.
Duan, F. K., Liu, X. D., Yu, T., et al.: Identification and estimate of biomass burning contribution to the urban aerosol organic carbon concentrations in Beijing, Atmos. Environ., 38, 1275–1282, 2004.
Fung, K. K., Chow, J. C., and Watson, J. G.: Evaluation of OC/EC speciation by thermal manganese dioxide oxidation and the IMPROVE method, J. Air Waste Manage. Assoc., 52 (11), 1333–1341, 2002.
Gao, Y., Arimoto, R., Duce, R. A., et al.: Temporal and spatial distributions of dust and its deposition to the China Sea, Tellus, Ser. B, 49, 172–189, 1997.
Gray, H. A., Cass, G. R., Huntzicker, J. J., et al.: Characteristics of atmospheric organic and elemental carbon particle concentrations in Los Angeles, Environ. Sci. Technol., 20 (6), 580–589, 1986.
Hansen, A. D. A., Bodhaine, B. A., Dutton, E. G., et al.: Aerosol black carbon measurements at the south pole: initial results, 1986–1987, Geophys. Res. Lett., 15, 1193–1196, 1988.
He, K. B., Yang, F. M., Ma, Y. L., et al.: The characteristics of $PM_{2.5}$ in Beijing, China, Atmos. Environ., 35, 4959–4970, 2001.

Hies, T., Treffeisen, R., Sebald, L., et al.: Spectral analysis of air pollutants, Part 1: elemental carbon time series, Atmos. Environ., 34, 3495–3502, 2000.
Holler, R., Tohnoa, S., Kasaharaa, M., et al.: Long-term characterization of carbonaceous aerosol in Uji, Japan, Atmos. Environ., 36, 1267–1275, 2002.
IPCC, Intergovernmental Panel on Climate Change, 2001: Climate Change 2001: The Scientific Basis, 5. Aerosols, their Direct and Indirect Effects, edited by: Houghton, J. T., Ding, Y., Griggs, D. J., Noguer, M., van der Linden, P. J., and Xiaosu, D., Cambridge University Press, London, 289–348, 2001.
Jacobson, M. Z.: Control of fossil-fuel particulate black carbon and organic matter, possibly the most effective method of slowing global warming, J. Geophys. Res., 107(D19), 4410, doi:10.1029/2001JD001376, 2002.
Kim, E., Hopke, P. K., and Edgerton, E. S.: Source identification of Atlanta aerosol by positive matrix factorization, J. Air & Waste Manage. Assoc., 53, 731–739, 2003a.
Kim, E., Hopke, P. K., and Edgerton, E. S.: Improving source identification of Atlanta aerosol using temperature resolved carbon fractions in positive matrix factorization, Atmos. Environ., 37, 5009–5021, 2003b.
Kim, E. and Hopke, P. K.: Improving source identification of fine particles in a rural northeastern U.S. area utilizing temperature-resolved carbon fractions, J. Geophys. Res., 109, D09204, doi:10.1029/2003JD004199, 2004.
Lee, H. S. and Kang, B. W.: Chemical characteristics of principal $PM_{2.5}$ species in Chongju, South Korea, Atmos. Environ., 35, 739–746, 2001.
Li, Y.: Characterization and source apportionment of atmospheric organic and elemental carbon in Xi'an, China, Master thesis of Institute of Earth Environment, (in Chinese), 2004.
Lin, J. J. and Tai, H. S.: Concentrations and distributions of carbonaceous species in ambient particles in Kaohsiung City, Taiwan, Atmos. Environ., 35, 2627–2636, 2001.
Menon, S., Hansen, J., Nazarenko, L., et al.: Climate effects of black carbon aerosols in China and India, Science, 297, 2250–2253, 2002.
Ohta, S., Hori, M., Yamagata, S., and Murao, N.: Chemical characterization of atmospheric fine particles in Sapporo with determination of water content, Atmos. Environ., 32(6), 1021–1025, 1998.
Pandis, S. N.: Secondary organic aerosol: precursors, composition, chemical mechanisms, and environmental conditions. Secondary Organic Aerosols Research Strategy to Apportion Biogenic/Anthropogenic Sources: An Outcome of the First Secondary Organic Aerosols

Workshop, February 4–5, 2002, Desert Research Institute, Reno, Nevada, Fort Lewis College, Durango, CO, 2002.
Schmid, H., Laskus, L., Abraham, H. J., et al.: Results of the "carbon conference" international aerosol carbon round robin test stage I, Atmos. Environ., 35, 2111–2121, 2001.
Streets, D. G., Gupta, S., Waldho, S. T., et al.: Black carbon emissions in China, Atmos. Environ., 35, 4281–4296, 2001.
Thurston, G. D. and Spengler, J. D.: A quantitative assessment of source contributions to inhalable particulate matter of pollution in metropolitan Boston, Atmos. Environ., 19, 9–25, 1985.
Turpin, B. J. and Huntzicker, J. J.: Secondary formation of organic aerosol in the Los Angeles Basin: a descriptive analysis of organic and elemental carbon concentrations, Atmos. Environ., 25A, 207–215, 1991.
Turpin, B. J. and Lim, H. J.: Species contributions to $PM_{2.5}$ mass concentrations: revisting common assumptions for estimating organic mass, Aerosol Sci. Technol., 35, 602–610, 2001.
Watson, J. G., Chow, J. C., and Houck, J. E.: $PM_{2.5}$ chemical source profiles for vehicle exhaust, vegetative burning, geological material, and coal burning in Northwestern Colorado during 1995, Chemosphere, 43, 1141–1151, 2001.
Watson, J. G., Chow, J. C., Lowenthal, D. H., et al.: Differences in the carbon composition of source profiles for diesel- and gasoline-powered vehicles, Atmos. Environ., 28, 2493–2505, 1994.
Wei, F., Teng, E., Wu, G., et al.: Ambient concentrations and elemental compositions of PM_{10} and $PM_{2.5}$ in four Chinese cities, Environ. Sci. Technol., 33, 4188–4193, 1999.
Yang, F., He, K., Ye, B., et al.: One-year record of organic and elemental carbon in fine particles in downtown Beijing and Shanghai, Atmos. Chem. Phys. Discuss., 5, 217–241, 2005, SRef-ID: 1680-7375/acpd/2005-5-217.
Ye, B. M., Jia, X. L., Yang, H. Z., et al.: Concentration and chemical composition of $PM_{2.5}$ in Shanghai for a 1-year period, Atmos. Environ., 37, 499–510, 2003.
Zhang, X. Y., Cao, J. J., Li, L. M., et al.: Characterization of Atmospheric Aerosol over XiAn in the South Margin of the Loess Plateau, China, Atmos. Environ., 36, 4189–4199, 2002.
Zhang, X. Y., Cao, J. J., Su, G. L., et al.: Particulate Pollution Control in Xi'an, in: United Nations Development Program & China International Center of Economic and Technology Exchange, Urban Air Pollution Control in China, China Science & Technology Press, (In Chinese), 222–293, 2001.

Zhang, X. Y., Arimoto, R., An, Z. S., et al.: Atmospheric trace elements over source regions for Chinese dust: Concentrations, sources and atmospheric deposition on the loess plateau, Atmos. Environ., 27(13), 2051–2067, 1993.
Zhuang, G. S., Zhen, Y., Duce, R. A., and Brown, P. R.: Link between iron and sulphur cycles suggested by detection of Fe (II) in remote marine aerosols, Nature, 355, 537–539, 1992.

Table 1. Average of the concentrations of OC and EC during September 2003 to February 2004 at Xi'an, China.

Season	Month	Sample numbers		OC[a]		EC		OC/EC	
		$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}	$PM_{2.5}$	PM_{10}
Fall	September	18	6	24.9±10.3[b]	29.6±11.2	8.3±4.5	11.0±6.4	3.3	3.0
	October	31	11	39.4±19.4	50.7±30.6	13.1±7.5	17.2±12.2	3.4	3.3
	Average	49	17	34.1±18.0	43.2±27.1	11.3±6.9	15.0±10.7	3.3	3.2
Winter	November	27	8	52.4±27.1	67.5±25.7	12.1±5.1	19.8±8.2	4.3	3.6
	December	29	8	81.7±36.2	124.8±54.8	15.2±4.6	28.9±8.9	5.3	4.3
	January	31	11	63.9±36.0	80.3±42.4	10.1±5.8	16.1±8.8	6.4	5.1
	February	29	9	48.6±21.7	98.7±87.6	12.0±4.4	26.8±18.2	4.1	3.5
	Average	116	36	61.9±33.2	93.0±58.4	12.3±5.3	22.7±12.3	5.1	4.2

Note: [a] unit is $\mu g\ m^{-3}$; [b] values represent average ± standard deviation

ACPD
5, 3561–3593, 2005

Table 2. Distribution of $PM_{2.5}$ mass, OC, EC, total carbonaceous aerosol (TCA)[a]% and OC/EC ratios under precipitation and non precipitation days.

Type	Sample numbers	$PM_{2.5}$ mass ($\mu g\ m^{-3}$)	OC($\mu g\ m^{-3}$)	EC($\mu g\ m^{-3}$)	TCA%	OC/EC
precipitation days						
Fall	13 (27%[b])	65.3 [c]	16.2	4.3	50.0	4.0
		(26.3–129.1)[d]	(9.5–28.2)	(1.7–9.0)	(35.2–80.9)	(3.1–6.2)
Winter	6 (5%)	128.4	34.7	8.6	50.6	4.0
		(74.5–168.7)	(19.0–49.6)	(6.8–11.9)	(38.1–57.7)	(2.8–5.3)
non precipitation days						
Fall	36 (73%)	165.0	40.2	13.7	48.9	3.1
		(51.2–327.6)	(16.4–74.2)	(3.3–27.6)	(30.9–69.8)	(2.2–4.9)
Winter	110 (95%)	250.0	63.3	12.5	45.7	5.1
		(40.4–663.9)	(9.9–189.6)	(2.2–25.1)	(23.7–72.9)	(2.8–9.4)

Note: [a] TCA=1.6×OC+EC; [b] the percentage in total observational days; [c] average; [d] range

ACPD
5, 3561–3593, 2005

Table 3. Statistical summary of the percentage of OC, EC and TCA% in $PM_{2.5}$ and PM_{10}.

Season	Month	TCA (%)			OC (%)			EC (%)		
		$PM_{2.5}$	PM_{10}	$PM_{2.5}/PM_{10}$	$PM_{2.5}$	PM_{10}	$PM_{2.5}/PM_{10}$	$PM_{2.5}$	PM_{10}	$PM_{2.5}/PM_{10}$
Fall	September	45.0±7.6 [a]	26.4±1.5	83.0±6.4	23.5±4.3	13.6±1.0	84.9±5.8	7.3±1.6	4.7±0.8	74.8±13.4
	October	51.0±10.7	38.8±8.8	79.4±6.2	26.6±5.5	20.4±4.8	80.2±6.1	8.4±2.7	6.3±1.6	75.0±10.1
	Average	48.8±10.1	34.5±9.3	80.7±6.3	25.5±5.3	18.0±5.1	81.8±6.3	8.0±2.4	5.7±1.6	75.0±11.0
Winter	November	44.8±7.4	35.7±4.6	69.8±11.8	24.3±3.9	18.9±2.5	71.8±12.2	5.9±1.7	5.4±0.9	58.7±12.0
	December	50.3±5.5	42.4±7.7	65.9±13.3	27.9±2.8	23.0±4.5	67.6±14.0	5.6±1.5	5.6±1.3	55.2±10.6
	January	44.0±8.1	37.0±11.7	75.0±9.1	24.9±4.6	20.6±7.1	77.0±9.6	4.0±1.1	4.1±0.8	61.4±7.3
	February	44.8±7.5	32.4±7.4	72.2±11.9	24.2±4.4	17.2±4.3	74.0±13.1	6.1±1.4	4.9±0.8	63.2±9.4
	Average	45.9±7.5	37.0±8.9	71.0±11.5	25.4±4.2	20.0±5.3	72.8±12.1	5.4±1.6	5.0±1.1	59.6±9.8

Note: [a] values represent average ± standard deviation

ACPD
5, 3561–3593, 2005

Table 4. Comparison of $PM_{2.5}$ OC, EC at Xi'an with other Asian cities.

City	Period	TC	OC	EC	OC/EC	Measurement method	Reference
		($\mu g\ m^{-3}$)					
Xi'an	Fall, 2003	45.4	34.1±18.0	11.3±6.9	3.3	IMPROVE-TOR	This study
	Winter, 2003	74.2	61.9±33.2	12.3±5.3	5.1	IMPROVE-TOR	This study
Beijing[1]	Fall, 1999	39.0	28.8	10.2	2.8	IMPROVE-TOR	He et al. (2001)
	Winter, 1999	42.6	31.5	11.1	2.8	IMPROVE-TOR	
Beijing[2]	2002.12	51.9	36.7±19.4	15.2±11.1	3.5	Elemental analyzer	Dan et al. (2004)
Beijing[3]	1997.11–1998.10		41.5 [4]	No data		Elemental analyzer	Duan et al. (2003)
Shanghai[5]	Fall, 1999	23.2	16.3	6.9	2.4	IMPROVE-TOR	Ye et al. (2003)
	Winter, 1999	25.1	17.0	8.1	2.1	IMPROVE-TOR	
Hong Kong	2002.1–2	14.4	9.6±4.5	4.7±2.9	2.3	IMPROVE-TOR	Cao et al. (2003)
Guangzhou	2002.1–2	31.0	22.6±18.0	8.3±5.6	2.7	IMPROVE-TOR	Cao et al. (2003)
Shenzhen	2002.1-2	19.2	13.2±4.1	6.1±1.8	2.2	IMPROVE-TOR	Cao et al. (2003)
Zhuhai	2002.1–2	17.3	12.2±4.4	5.0±1.6	2.4	IMPROVE-TOR	Cao et al. (2003)
Kaohsiung	1998.11–1999.4	14.5	10.4	4.0	2.6	Elemental analyzer	Lin and Tai (2001)
Chongju, Korea	Fall, 1995	12.4	6.0	6.4	0.9	IMPROVE-TOR	Lee and Kang (2001)
	Winter, 1995	9.3	5.0	4.3	1.2	IMPROVE-TOR	
Sapporo, Japan	1992.9–10	9.1	4.1	5.0	0.8	Elemental analyzer	Ohta et al. (1998)
	1992.1–2	9.0	3.9	5.1	0.8	Elemental analyzer	
Uji, Japan	1998.9–10	6.4	1.8	4.6	0.4	R&P 5400	Holler et al. (2002)
	1998.11–12	10.2	2.5	7.7	0.3	R&P 5400	

Note: [1] Chegongzhuang site; [2] average of 3 sites: Beijing Normal University, Capital Steel Plant, Yihai Garden; [3] Temple of Heaven; [4] annual averagel [5] average of Tongji University and Hainan Road

ACPD
5, 3561–3593, 2005

Table 5. PCA results of fall Samples.

	PC1	PC2	PC3
OC1	0.32	0.12	0.91
OC2	0.96	0.16	0.17
OC3	0.89	0.11	0.38
OC4	0.95	0.19	0.19
OP	0.88	0.25	0.13
EC1	0.88	0.21	0.39
EC2	0.62	0.65	−0.28
EC3	0.12	0.94	0.23
Variance	68%	14%	10%
Eigenvalue	5.60	1.10	0.80
	Gasoline Exhaust	Diesel Exhaust	Biomass Burning

Table 6. PCA results of winter samples.

	PC1	PC2	PC3
OC1	0.09	0.98	0.06
OC2	0.97	0.03	0.05
OC3	0.87	0.44	0.07
OC4	0.96	0.05	−0.01
OP	0.60	0.67	0.24
EC1	0.73	0.50	0.17
EC2	−0.05	0.12	0.90
EC3	0.17	0.06	0.88
Variance	55%	21%	13%
Eigenvalue	4.40	1.60	1.10
	Coal combustion + Gasoline exhaust	Biomass burning	Diesel exhaust

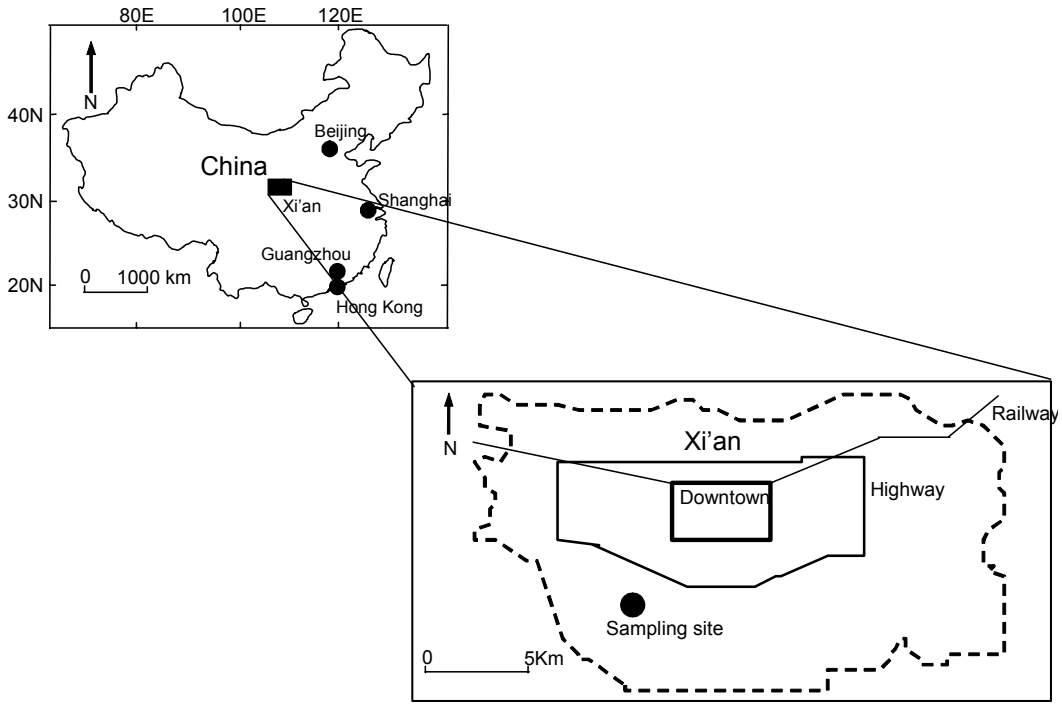

Fig. 1. Location of the sampling site at Xi'an, China.

ACPD
5, 3561–3593, 2005

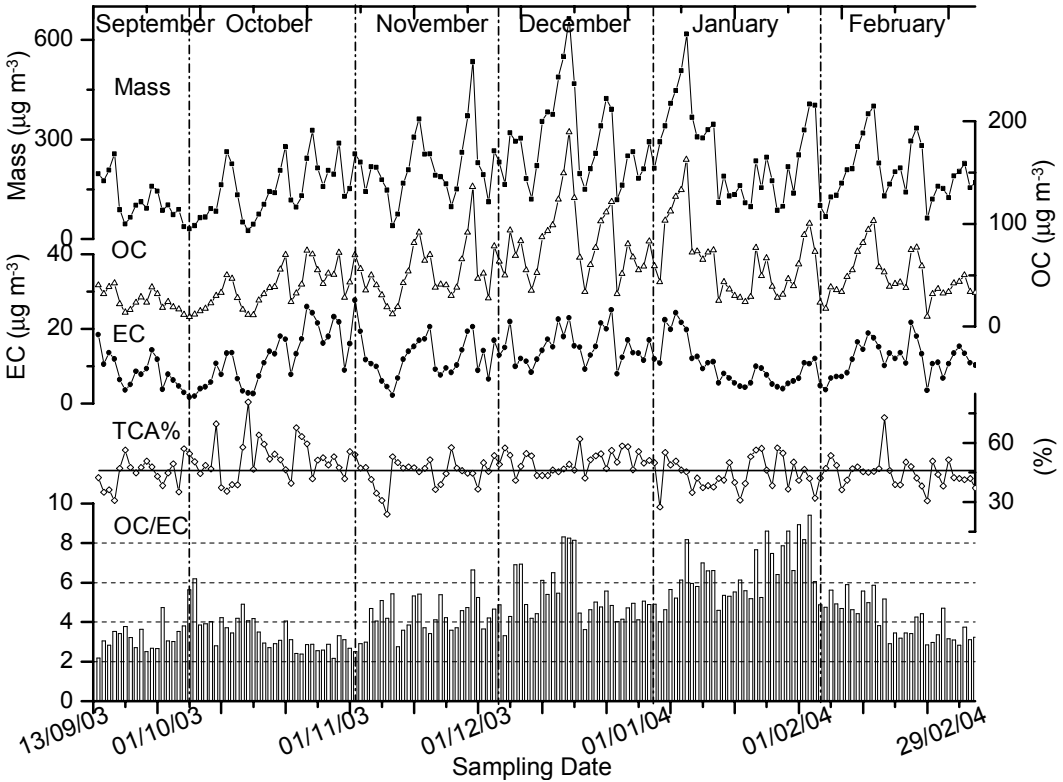

Fig. 2. Time series of $PM_{2.5}$ Mass, organic carbon (OC), elemental carbon (EC), TCA%, and OC/EC ratios at Xi'an from 13 September 2003 to 29 February 2004 (TCA is total carbonaceous aerosol, sum of organic matter (1.6×OC, Turpin and Lim, 2001) and EC).

ACPD
5, 3561–3593, 2005

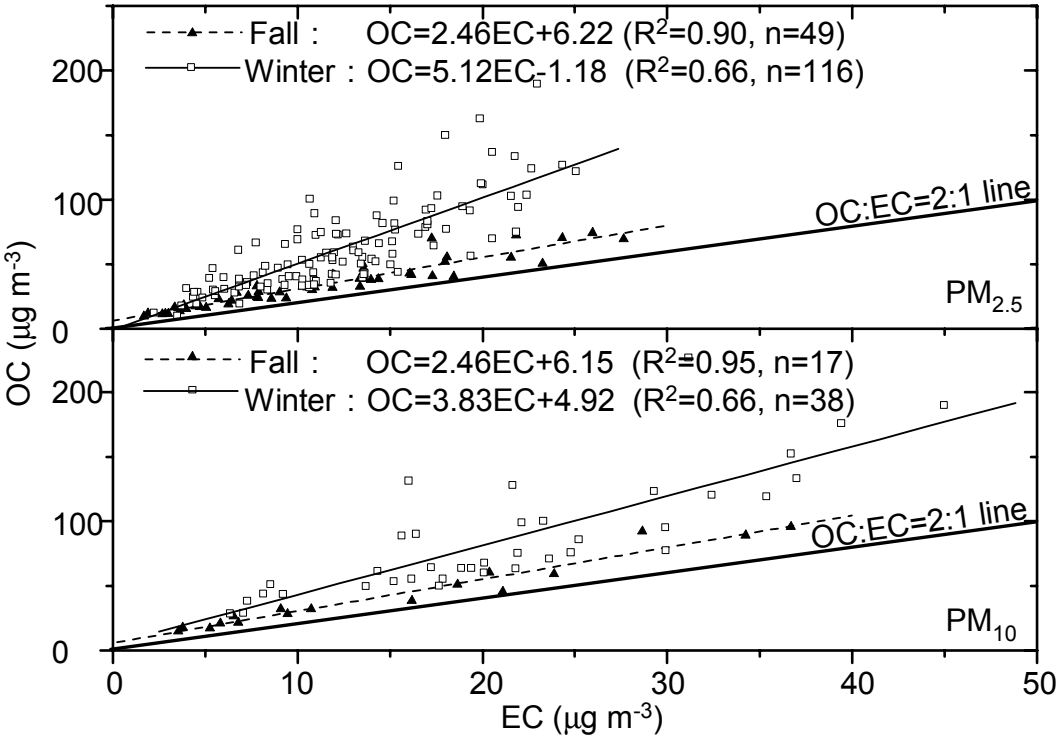

Fig. 3. Relationship between OC and EC concentrations in $PM_{2.5}$ and PM_{10}.

ACPD
5, 3561–3593, 2005

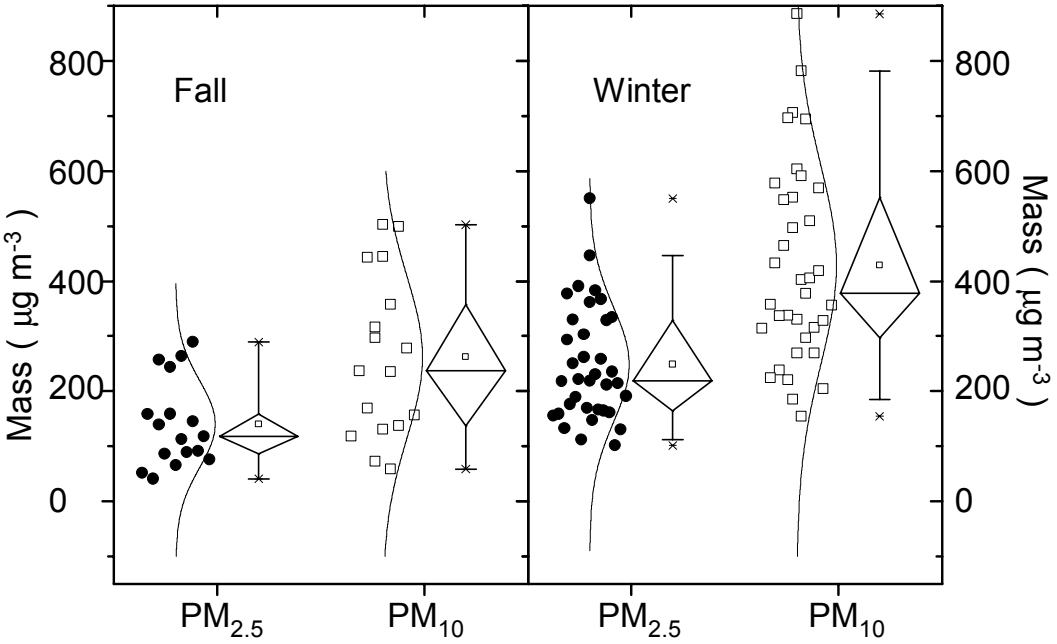

Fig. 4. Distribution of $PM_{2.5}$ and PM_{10} mass concentrations during fall and winter (The valid paired samples were 17 in fall and 36 in winter. The points in the figure were measured data and the curves were the normal fitting curve of these data.)

ACPD
5, 3561–3593, 2005

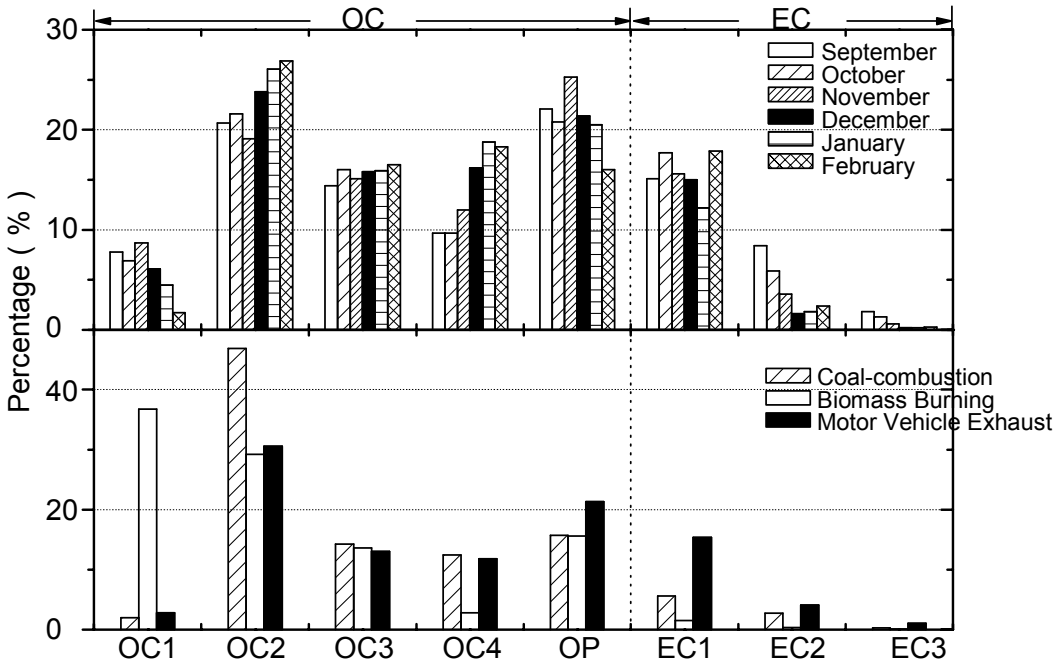

Fig. 5. Percentage of total carbon contributed by eight carbon fractions in ambient air and three major source samples.

ACPD
5, 3561–3593, 2005

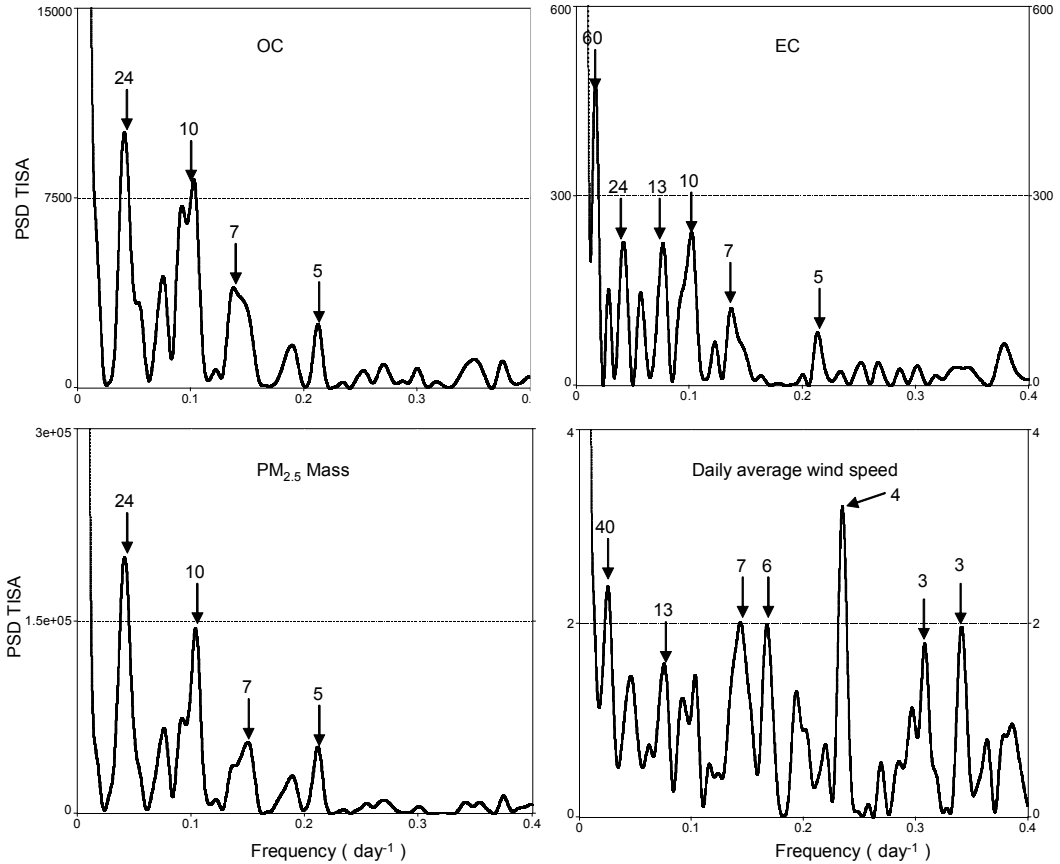

Fig. 6. Periodicity of $PM_{2.5}$ OC, EC, mass, and daily average wind speed. (PSD TISA refers to Power as Time-Integral Squared Amplitude.)

ACPD
5, 3561–3593, 2005

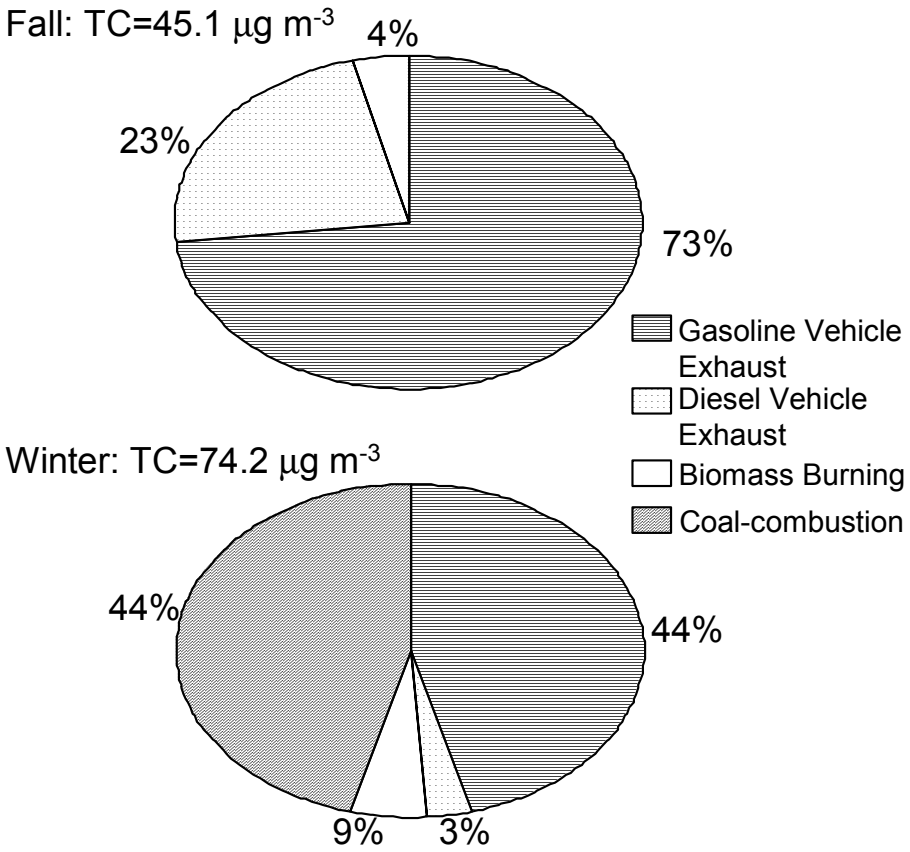

Fig. 7. Relative contributions of major sources to $PM_{2.5}$ TC during fall and winter, 2003.